

# **METHODS AND APPARATUS FOR HIGH-SHEAR MIXING AND REACTING OF MATERIALS**

**By**

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## **CROSS-REFERENCE TO RELATED APPLICATIONS**

**[0001]** This application claims the benefit of the prior filing date of U.S. provisional patent application number 60/410,185, filed September 11, 2002, herein incorporated by reference in its entirety.

## **BACKGROUND OF THE INVENTION**

### **FIELD OF THE INVENTION**

**[0002]** The invention is concerned with methods and apparatus for high-shear mixing and reacting of materials involving a chemical and/or a physical action(s) or reaction(s) of a component or between components.

### **REVIEW OF THE PRIOR ART**

**[0003]** Apparatus for materials processing consisting of coaxial cylinders that are rotated relative to one another about a common axis, the materials to be processed being fed into the annular space between the cylinders, are known, as shown for example, in US Patent No. 5,370,999, issued 6 December 1994 to Colorado State University Research Foundation, and US Patent No. 5,430,891, issued 23 August 1994 to Nippon Paint Co., Ltd.. My US Patents Nos. 5,279,463 (issued 18 January, 1994); 5,538,191 (issued 23 July 1996); and pending US application No. 09/802,037 (filed March 7, 2001) disclose methods and apparatus for high-shear material treatment, one type of the apparatus consisting of a rotor rotating within a stator to provide an annular flow passage comprising a flow path of uniform radial dimension along its length containing a high-shear treatment zone in which free supra-Kolmogoroff eddies are suppressed during passage of the material therethrough. In another type of the apparatus the passage spacing at one location on its circumference is smaller than in the remainder of the zone to provide a subsidiary higher-shear treatment zone in which free supra-Kolmogoroff eddies are suppressed.

**[0004]** Couette developed an apparatus for measuring the viscosity of a liquid consisting of a cylinder immersed in the liquid contained in a rotating cylindrical vessel, the viscosity being measured by measuring the torque that was thus applied to the cylinder.

It was found that a linear relationship was obtained between the viscosity measurement and the angular velocity of the rotor surface up to a certain value beyond which the linear relationship broke down. This phenomenon was investigated by G.I. Taylor who showed that when a certain Reynolds number was exceeded the previously stratified flow in the annulus between the two cylindrical surfaces became unstable and vortices appeared, now known as Taylor vortices, whose axes are located along the circumference of the rotor parallel to its axis of rotation and which rotate in alternately opposite directions. The conditions for the flow to become unstable in this manner can be expressed with the aid of a characteristic number known as the Taylor number, depending upon the radial width of the annular gap, the radius of the rotor and its peripheral velocity. I have found, as described in my pending US application No. 09/802,037 (filed March 7, 2001), the disclosure of which is incorporated herein by this reference, that when using such apparatus for the types of process where thorough mixing is required, the presence of the Taylor vortices inhibits the action or reaction desired since the material to be treated becomes partially entrapped in the vortices, whereupon mixing is impaired and is required to be replaced by much slower diffusion processes.

#### **BRIEF SUMMARY OF THE INVENTION**

**[0005]** It is a principal object of the invention to provide new methods and apparatus for high-shear mixing and reacting of materials wherein interdiffusion mixing can be obtained rather than macroscopic convection mixing, and the apparatus employed can be operated without the presence of Taylor vortices so that such interdiffusion mixing is obtained.

**[0006]** It is another more specific object to provide new methods and apparatus for high-shear mixing and reacting of materials wherein interdiffusion mixing can be obtained rather than macroscopic convection mixing without requiring the use of relatively moving surfaces in the treatment passage.

**[0007]** In accordance with the invention there is provided a new method for high-shear mixing and reacting of materials comprising:

supplying a first material to a flow path through an interdiffusion passage between two parallel, closely spaced, stationary surfaces at a respective first flow rate;

supplying a second material to the flow path through the interdiffusion passage at a respective second flow rate, to be interdiffused with the first material therein, with resultant material from the interdiffusion and any consequent reaction moving in the flow path at a respective resultant flow rate;

wherein the first and the second materials, and material resulting from interdiffusion and any consequent reaction of the materials, form respective boundary layers against both surfaces;

wherein the radial spacing between the two parallel surfaces is equal to or less than the back-to-back radial thicknesses of the two laminar boundary layers of material against the two surfaces, and if larger than the back-to-back radial thicknesses with a third layer between the two boundary layers has the third layer too thin to support turbulent convection or uneven channeling; and

wherein the flow rates of the materials in the flow path are such that they are subjected to laminar shear of the value required for the interdiffusion.

**[0008]** Also in accordance with the invention there is provided new apparatus for high-shear mixing and reacting of materials comprising:

apparatus structure providing two closely spaced stationary surfaces constituting between them a flow path that is an interdiffusion passage;

means supplying a first material to the flow path through the interdiffusion passage at a respective first flow rate;

means supplying a second material to the flow path through the interdiffusion passage at a respective second flow rate, to be interdiffused with the first material therein, with resultant material from the interdiffusion and any consequent reaction moving in the flow path at a respective resultant flow rate;

wherein the first and the second materials, and material resulting from interdiffusion and any consequent reaction of the materials, form respective laminar boundary layers against both surfaces;

wherein the radial spacing between the two parallel surfaces is equal to or less than the back-to-back radial thicknesses of the two boundary layers of the material against the two surfaces, and if larger than the back-to-back radial thicknesses with a third layer between the two boundary layers has the third layer too thin to support turbulent convection or uneven channeling; and

wherein the means supplying the first and second materials supply those materials at flow rates such that the materials in the passage are subjected to laminar shear of the value required for the interdiffusion.

**[0009]** Preferably, the two parallel spaced surfaces are provided by two cylindrical apparatus members mounted one within the other, so that the inner surface of the outer member and the outer surface of the inner member constitute two parallel, closely spaced smooth surfaces providing an annular interdiffusion passage constituting the flow path for the materials.

#### **DESCRIPTION OF THE DRAWINGS**

**[0010]** Methods and apparatus that are particular preferred embodiments of the invention will now be described, by way of example, with reference to the accompanying diagrammatic drawings.

**[0011]** Figure 1 is a longitudinal cross section through the apparatus.

**[0012]** Figure 2 is a cross sectional view of a portion of the plenum depicting a flow control mechanism.

#### **LIST OF REFERENCE NUMBERS IN DRAWINGS**

- 10 Cylindrical outer casing
- 12 Longitudinal axis of casing 10 and inner body 18
- 14 Circumferential plenum to outer casing 10
- 16 Circumferential inlet from plenum 14
- 18, 18A, 18B, 18C Hollow inner body and parts thereof
- 20 Radial support struts between bodies 10 and 18
- 22, 22A, 22B, 22C Processing passage and portions thereof
- 24 Cylindrical inlet to passage 22
- 26 Side inlet to plenum 14
- 28 Outlet from passage 22
- 30 Inner cylindrical surface of outer member 10

- 32 Outer cylindrical surface of inner body 18
- 34 Heat exchanger surrounding outer member 10
- 36 Receptor plate at outlet 28
- 38 Heater for receptor plate 36
- 40 Frusto-conical passage receiving discharge from outlet 28
- 42 Structures forming passage 40
- 44 Heaters for passage 40
- 46 Toroidal shroud plate surrounding passage 40
- 48 Gas outlet from upper end of shroud 46
- 50 Trough at lower end of shroud 46
- 52 Liquid outlet from trough 50
- 54 Mist eliminator in passage 40
- 56 Roof
- 58 Upper flap
- 58A Upper flap contact portion
- 60 Lower flap
- 60A Lower flap contact portion

#### **DETAILED DESCRIPTION OF THE INVENTION**

[0013] The apparatus illustrated by Figure 1 comprises a cylindrical outer casing 10 symmetrical about a longitudinal axis 12, the casing having an outward tapering inlet portion 10A connected with a central portion 10B of uniform diameter along its length, the central portion in turn being connected with inward tapering exit portion 10C. The casing 10 is formed with a circumferential plenum 14 having a circumferential inlet 16 to the interior of the casing 10. A hollow inner body 18 of approximately the same shape as the interior of the casing 10, but somewhat smaller in diameter, is mounted within the casing, as by streamline shaped radial struts 20, so as to have its longitudinal axis coincident with that of the casing 10. One example of such a radial strut is a set screw. The number and spacing of these radial struts 20 is dependent on the operating conditions and sizing of the components of the apparatus. Thus, the inner body has an inlet portion 18A, a central portion 18B and an exit portion 18C, so as to form between its exterior surface and the interior surface of the casing an annular cross section processing passage having an inlet portion 22A, a central portion 22B and an exit portion 22C.

**[0014]**The circumferential inlet 16 may be fitted with a flow control mechanism which allows the passage of material from plenum 14 in only one direction, namely, into the processing passage 22B, and blocks or restricts flow from the processing passage into the plenum. Depending on the reactions occurring in the apparatus, including the relative pressures of the reactants in passage 22 and plenum 14, a flow control mechanism may or may not be required. Figure 1 shows a thin line at inlet 16, which indicates a very simple, thin, spring-like circumferential flap, which operates as a check valve. The material should preferably be flexible and nonreactive, such as stainless steel, or a Teflon like elastomer, one example being sold under the brand name KALREZ. Another version of such a flow control mechanism is depicted in Figure 2. Figure 2 also shows an internal shape of plenum 14 having a sloping roof 56. The shape of plenum 14 is a design expedient, and many shapes would work without impacting the invention described and claimed herein. Attached to roof 56 and extending into processing passage 22B is upper flap 58. Cooperating with upper flap 58 is lower flap 60, which is attached to the interior wall of plenum 14. When the pressure of reactant B is less than the pressure in passage 22, portions of flaps 58 and 60 are in contact with one another, namely contact portions 58A and 60A, and block or restrict flow of material from passage 22 into plenum 14. When the pressure of reactant B exceeds the pressure in passage 22, then the contact portions 58A and 60A separate, and reactant B flows into the passage 22B.

**[0015]**A first material to be processed in the passage is supplied to the passage inlet portion 22A through a cylindrical inlet 24 disposed with its longitudinal axis coincident with the axis 12 so that the material is fed uniformly into the annular passage. In the event that the material is a liquid or slurry, or otherwise pumpable, then it will be supplied under the urge of a pressure pump of sufficient capacity to feed the material into the passage at a corresponding required first flow rate; if on the other hand the material is a gas it can instead be supplied from a pressurized reservoir thereof through a pressure controlling valve. Such means for the supply of the material are known to those skilled in the art and do not require specific illustration herein. A second material to be processed in the passage is supplied to the junction of the passage inlet portion 22A and passage central portion 22B via a side inlet 26 feeding into the plenum 14, so

that the material is fed uniformly into the annular passage and into the stream of the first material passing therein. Again, as with the first material, in the event that it is a liquid or slurry, or otherwise pumpable, then it will be supplied under the urge of a pressure pump of sufficient capacity to feed the material into the passage at a corresponding required second flow rate; if on the other hand the material is a gas it can instead be supplied from a pressurized reservoir thereof through a pressure controlling valve. The interdiffused, and possibly reacted, material that passes through the outlet portion 22C of the annular passage is discharged through an outlet 28.

**[0016]**As the material being processed flows through the processing passage a respective laminar boundary layer forms on each of the inner cylindrical surface 30 of the outer member 10 and the outer cylindrical surface 32 of the inner member 18, the thickness of which is determined by the viscosity and other factors of the material being processed and the flow velocity of the material over the surface. The thickness of such a laminar boundary layer for any fluid may be determined by the Blasius equation, which takes account of the path length of the flow, the flow velocity and the kinematic velocity. The internal diameter of the surface 30 and the external diameter of the surface 32 at the respective central portions 10B and 18B are such that the radial dimension of this portion 22B of the processing passage is just equal to the combined thicknesses of the two boundary layers back-to-back on these surfaces, or is so close to this value that there is no room between them for an intervening bulk layer thick enough to permit turbulent macro-agitation to be formed and disrupt the thorough interdiffusion that takes place of the materials in the two contacting laminar boundary layers. It has been discovered that such interdiffusion of highly sheared laminar boundary layers effects a significant increase in the speed and uniformity of molecular diffusion as is desired for very fast chemical reaction to take place. As a specific example, with apparatus in which the inner body 18 is of 7.5cm external diameter a gap of 1-12mm is likely to be required to ensure the absence of any bulk layer between the boundary layers of liquids having dynamic viscosities of 1.0 cP or higher; if the gap is increased to as little as 5mm the resulting intervening turbulent bulk layer is almost certain to slow down the otherwise very fast interdiffusion and subsequent fast chemical reactions.

[0017] It is essential in order to ensure the required degree of interdiffusion while the materials are resident in the passage to maintain a minimum flow velocity in order to maintain the desired high shear interaction between the boundary layers. This velocity should not be less than about 5.0 meters per second. It is also desirable that the surfaces 30 and 32 should be of a certain degree of smoothness and inertness in order to prevent undue retention of liquid residues thereon. The normal machining of the cylindrical surfaces to the tolerances required for the parameters indicated above (e.g. a radial gap of 1-12mm for a rotor 7.5cm diameter) is insufficient and the resultant roughness of the surface, although not normally regarded as roughness, is still sufficient to allow thin deposits to form, and it is found desirable therefore to polish these surfaces to a so-called mirror finish, or better. The standard finish of 40 microinches is definitely too coarse, and a honed finish to at least 10 microinches, and preferably 5 microinches, is preferred.

[0018] The methods and apparatus of the invention are operable, for example, to quickly forcibly dissolve gases in liquids in which they are normally of low solubility, or to virtually instantaneously emulsify non-miscible liquids, or to chemically react two or more materials together with very high reaction rates, sometimes even in the absence of catalysts, surface active materials, etc., that frequently are required in conventional processes to obtain economically acceptable reaction rates. Some of the processes to which they are applicable will involve only physical interdiffusion of the materials fed into the apparatus, e.g. emulsification, while others will involve chemical reaction with or without the possibility of simultaneous physical interaction. In a number of the processes one of the materials fed, for example, into the passage 22 via the inlet 24 (the first material) may comprise a mixture of materials which will interdiffuse and interact in the desired manner in its journey through the passage, while the material fed in through the inlet 26 (the second material) is an inert gas under high pressure, e.g. nitrogen or air, whose sole purpose is to produce an adequate flow rate of the first material through the passage. Many chemical reactions result in the production of gaseous products at the temperature and pressure of the reaction, and in the confined space of the processing passage this may be sufficient to produce the required accelerated flow rate through the passage. It will be noted that when such production

does occur it will be necessary for the materials to be fed into the passage at a sufficiently high pressure so that the internal pressure generated does not cause blow-back of the materials.

**[0019]** In general, most chemical reactions and many physical reactions are to a greater or lesser degree either endothermic or exothermic, and many are quite strongly so. The higher reaction rates that are possible result in a corresponding considerably increased production or loss of heat, some of which can be transferred out of the apparatus via the exiting fluid/s, but the remainder of which must be transferred though the walls of the casing and/or inner body if the process temperature is to be maintained within required limits. Another factor that is important in such apparatus is that the heat conductivity of the two thin boundary layer films is very high, since there is no bulk layer between them through which the heat must pass, as with conventional bulk stirring systems. The achievement of the highest possible heat transfer rate, if possible higher than is strictly necessary in order to provide a margin for adjustment, is therefore desirable to ensure that the processing temperature can at all times readily be maintained within those required limits, which can constitute a very narrow range, e.g. 1°C. To such end the apparatus is provided with a heat exchanger structure 34 fitting snugly around the exterior surface of the outer body 10. This may be of any conventional known type, but preferably is an impingement heat exchanger as described and claimed in my U.S. application No. 60/318,985, filed September 13, 2001 the disclosure of which is incorporated herein by this reference.

**[0020]** The apparatus specifically described is intended for operation with a process in which a copious quantity of gaseous material is a by-product of the reaction, with the result that a jet of the reacted material, consisting of a mixture of gas, vapor and liquid is delivered forcefully out through the outlet 28. The jet impinges on a receptor plate 36 heated by a heater 38, which diverts the stream radially outward to enter a frusto-conical shaped passage 40 formed between two correspondingly shaped structures 42.

These structures are provided with a series of heaters 44 which are controlled to maintain the temperature of the passage 40 at a desired value. The plate 36 and structures 42 are surrounded by a toroidal shroud 46 having a gas outlet 48 at its upper end and providing a trough 50 at its lower end into which liquid in the discharge from the

passage 22 that deposits on the lower structure 42 can drain and exit from the apparatus via an outlet 52. Vapor mist in the passage discharge decreases in velocity as it moves radially outward and will coalesce and also deposit on the lower frustoconical structure 42 to drain into the trough 50; a mist eliminator 54, consisting for example of an conical annulus filling body of expanded metal, is disposed in the passage 40 to ensure that all the vapor is separated from the purely gaseous component.

[0021] The methods and apparatus of the invention therefore provide for interdiffusion of the materials fed to the processing passage for processes in which this is possible, without the need for relative rotation between the bodies 10 and 18, as is required with the processes and apparatus described in my pending US application No. 09/802,037 (filed March 7, 2001), considerably simplifying the apparatus and reducing its primary cost and cost of operation by the elimination of a drive motor, supporting bearings, etc.. Such savings may however be offset by the need for higher operating pressures to ensure that the materials are subjected to laminar shear of the required values as a result of their flow through the processing passage, and to ensure that fouling and obstruction of the passage does not take place.

[0022] In the embodiment illustrated the longitudinal axes of the two bodies 10 and 18 are coincident so that the passage 22 is of uniform radial dimension about its circumference. However, instead the two axes can be parallel and displaced so as to provide an interdiffusion mixing zone which is of cyclically variable radial dimension. Again, although in the embodiment particularly illustrated the bodies 10 and 18 are cylindrical and disposed one within the other, in other embodiments they can be flat and parallel to provide a passage of correspondingly flat shape between them. The cylindrical cross section structure has the advantage however that, with radially outward operating pressures acting against a circular structure, it is less difficult to maintain the very small radial gap forming the passage 22 to the very close tolerance required under the high pressures that could be required for successful continuous operation, whereas a flat structure is likely subject to warping and change of the passage dimensions under such conditions.